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Swelling Resistance Induced by Grain Refinement and Particle Dispersion in Austenitic Stainless Steel during High Energy Electron Irradiation

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Abstract

Thin-foils of an austenitic stainless steel with and without dispersions of aluminum oxide particles are irradiated in a High Voltage Electron Microscope at 600°C; prior to irradiation, up to 1000 ppm of helium atoms are implanted in the thin foils. The void concentration and the swelling decreased with decreasing size of the grains irradiated; the effect of grain size being more marked in the dispersed specimens. The grain size effect persisted also in specimens containing high concentrations of helium. The influence of high concentrations of helium on void growth and swelling is investigated. The dose dependencies of void size and swelling for grains of different sizes and with different helium concentrations are described.

These results are analysed in terms of grain size dependent vacancy supersaturation and are found to be consistent with the "defect depletion" model recently proposed by the author. It is concluded that void nucleation is critically dependent on vacancy supersaturation and that the grain size effect is predominantly nucleation controlled.

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CONTENTS

1. INTRODUCTION

It has become apparent both from theoretical and experimental studies that the phenomenon of irradiation induced void formation is extremely complex and is strongly influenced by a large number of material variables. A better understanding of the effect(s) of these material variables on the formation and growth of voids is particularly desirable because of its direct relevance to the problem of designing a damage resistant material for fast breeder reactors and possibly also for fusion reactors.

The effects of some of these variables on the voidage behavior of metals and alloys have been the subject of several theoretical and experimental studies [1-4]. Since grain boundaries act a neutral sinks for both vacancies and self-interstitial at produced during irradiation, it is expected that the presence of grain boundaries would influence the swelling behavior of the material under irradiation [5]. A detailed and systematic study of the effect of grain size on void volume swelling has recently been made by the author [6-8]. It has also been demonstrated that the presence of grain boundaries strongly affects the growth behavior of voids situated immediately adjacent to the oundaries [9]. The grain size effect has been explained in terms of a "defect depletion" model [8]; our calculations of vacancy supersaturations in three-dimensional grains of different sizes [10] also support the model.

We have extended our study of grain size effect on void formation by investigating the effects of helium concentrations on the nucleation and growth of voids in grains of different sizes; this is described and discussed in the present paper. First, the effects of helium concentration and vacancy supersaturation on void nucleation are examined. This is followed by the analysis of the dose dependencies of void growth and void volume swelling in terms of vacancy supersaturation and helium concentration. Finally, the influence of very high concentrations of implanted helium atoms on void growth and void volume swelling is described and briefly discussed.

2. MATERIAL AND EXPERIMENTAL PROCEDURE

The materials used in the present investigation were manufactured, using powder metallurgical techniques, from pre-alloyed austenitic stainless steel powder of composition quoted in Table 1.

Table 1

Composition of pre-alloyed powder

Element	Fe	Ni	Cr	C	Mo	Nb	W	Si
Concentration (wt.%)	Bal.	20	20	0.02	ND*	ND*	ND*	0.22

* Not detectable

The finished products were obtained either in the form of extruded bar or extruded and hot-rolled sheet. The specimens are classified according to their composition, fabrication and heat treatment as shown in Table 2. All heat treatments were for two

Table 2

Specimen description

Designation	Description
RMS-1a	Composition as in Table 1, extruded, annealed at 800°C
RMS-1b	Same as RMS-1a, but annealed at 1150°C
RMS-1c	Composition as in Table 1, extruded and hot-rolled, annealed at 1150°C
RMS-2a	Composition as in Table 1, 1.0 wt.% Ti (powder), 5.0 v.% Al_2O_3 particles of 500 Å diameter (i.e. 0.76×10^{15} particles/cm^3), extruded and annealed at 800°C
RMS-2b	Same as RMS-2a but no Ti, extruded and hot-rolled, annealed at 800°C

hours under a vacuum of 10^{-6} torr. Thin films were prepared from the annealed samples by electro-polishing in ethanol with 20 v.% perchloric acid at -20°C.

In selected thin films helium atoms (up to 1000 ppm) were implanted at room temperature using a 100 kV Heavy Ion Accelerator at A.E.R.E. Harwell. Thin films, both with and without helium were irradiated at 1 MeV in the EM-7 Electron Microscope at Harwell; all irradiation experiments were carried out at 600°C. During irradiation a vacuum of $\sim 2\text{-}3 \times 10^{-7}$ torr was maintained around the specimens. The microscope current was adjusted to give a displacement rate of 5.16×10^{-3} dpa (displacement per atom) sec^{-1} within a 0.5 μm diameter circle on the specimen; the displacement rate was calculated according to the procedure outlined by Makin [11-12]. For comparison purposes, also a specimen of cast and solution-treated (at 1050°C for 30 min) AISI type 316 stainless steel was irradiated under exactly the same conditions as our RMS-1 and RMS-2 stainless steels. The average grain size of the 316 stainless steel specimens was of the order of 50 μm.

During irradiation the specimen structure was photographed at various stages. The void concentration, growth, and volume swelling data were obtained from these micrographs as described in ref. [8].

3. RESULTS

3.1. Nucleation of Voids and their Concentration

As reported earlier [8], the void concentration initially increases with the displacement dose and reaches a maximum C_v^{max}. The quantity C_v^{max} can be taken to represent the upper limit of the scale of nucleation for a given helium concentration and grain size. The variation of C_v^{max} with the size of the grain, d_g, and the helium concentration can, therefore, be used to study the effect of these parameters on the nucleation of voids. This is illustrated by the results quoted in Table 3; the average void size d_v at C_v^{max} is also quoted. The parameter $(S/S_o)^{max}$ mentioned in Table 3 is the steady-state vacancy supersaturation relative to a very large grain and refers to its value in the centre of a grain of diameter d_g. It should be noted here that for some grains in Table 3 d_g is quoted to be

Table 3

Maximum Void Concentration

Specimen	ϕt (dpa)	d_g (μm)	$(S/S_o)^{max}$	C_v^{max} (cm^{-3})	d_v (Å)	Helium content (ppm)
RMS-1c	13.95	>2.50	1.0	0.46×10^{15}	492	0
RMS-1c	9.30	>2.50	1.0	10.40 "	170	100
RMS-1c	15.50	>2.50	1.0	32.60 "	175	1000
RMS-1a	13.95	1.15	0.85	2.44 "	316	10
RMS-2a	7.44	1.06	0.81	5.53 "	175	100
RMS-2a	18.60	0.75	0.62	1.87 "	229	10
RMS-1a	18.60	0.70	0.58	2.51 "	340	10
RMS-2a	10.85	0.70	0.58	2.75 "	250	1000
RMS-2a	23.25	0.56	0.42	1.51 "	333	50
RMS-2b	26.97	0.52	0.38	0.71 "	330	10

> 2.5 μm. The reason for this is that in cases of large grains only a part of the grain under irradiation could be accomodated on the photographic film; in these large grains the size of the exposed part of the grain is > 2.5 μm.

It can be seen from data quoted in Table 3 that C_v^{max} is strongly dependent on the size of the grain under irradiation at all levels of helium content investigated. The effect of helium concentration on the nucleation of voids is also quite clear; for a given grain size, the void concentration increases with increasing helium concentration. At low helium concentration (i.e. 10 ppm) the dose at which C_v^{max} is reached generally decreases with increasing grain size. In the large grains (d_g > 2.50 μm) with 100 and 1000 ppm of helium, on the other hand, the dose at which C_v^{max} is reached tends to increase with increasing C_v^{max}. In small grains (d_g < 0.75 μm) the dose at which C_v^{max} is reached decreases with increasing C_v^{max} (i.e. increasing helium concentration).

3.2. Void Growth and its Dose Dependence

The influence of grain size and helium content on the variation of the average void size with square root of displacement dose both in undispersed and dispersed specimens are shown in Figs. 1 and 2, respectively. The effect of void concentration on the growth of voids is clear from Fig. 1; in large grains (d_g > 2.5 μm) the growth rate decreases with increasing void number density (the maximum void number density C_v^{max} for grains quoted in Figs. 1 and 2 are given in Table 3).

It can be seen from Figs. 1 and 2 that in order to discern the effect of the size of the irradiated grain on the growth rate of voids, it is essential to separate out the influence of void concentration on void growth. Bearing this in mind, the effect of grain size on the void growth can be illustrated by the following comparisons: (a) compare curve 1 (Fig. 1) with curve 2 (Fig. 2). Since the void concentrations are almost the same in both grains and since neither of the grains contain implanted helium, the lower void growth rate in the 0.45 μm grain (curve 2, Fig. 2) is in all probability due to smaller grain size: (b) Compare curve 3 (Fig. 1) with curve 4 (Fig. 2). It can be seen that although the void concentration for curve 4 (Fig. 2) is much lower than that for curve 3 (Fig. 1), the growth rates are very similar in both cases. This also suggests that the growth rate has been affected by the size of the grain:

(c) Compare curve 2 (Fig. 1) with curve 1 (Fig. 2). The growth rates are very similar in the two cases. This seems reasonable since the grains are of the same size and have very similar void concentrations. It should be noted however that the 0.70 μm grain with 10 ppm helium does not contain particles whereas the 0.70 μm grain with 1000 ppm helium is in a specimen containing dispersions of particles. It is significant that the void growth rate in the 0.70 μm grain (curve 1, Fig. 2) with 1000 ppm helium is higher than in the 0.75 μm grain (curve 4, Fig. 2) containing only 10 ppm helium although the void concentration is lower in the 0.75 μm grain than in the 0.70 μm grain; both grains are from dispersed specimens.

3.3. Void Volume Swelling and its Dose Dependence

The variation of void volume swelling (Δv/v) with (displacement dose)$^{3/2}$ for grains of different sizes and with different helium contents is shown in Fig. 3; the void volume per unit volume of the specimen, (Δv/v), refers to the swelling measured in the central region of each grain. It is quite clear from Fig. 3 that the void volume swelling is strongly dependent on the grain size, d_g when d_g < 2.50 μm. In grains with d_g > 2.50 μm, the effect of implanted helium on the void volume swelling is hardly noticeable except in the case of the grain containing 1000 ppm of helium. The swelling rate in the 0.70 μm grain containing 1000 ppm of helium is considerably higher than in the 0.75 μm grain containing only 10 ppm of helium (note that both grains are from dispersed specimens); the swelling rate in the former, however, is almost exactly the same as in the 0.70 μm undispersed grain containing only 10 ppm helium.

Results quoted in Fig. 3 also suggest that the void volume swelling for a given grain size containing 10 ppm of helium is significantly higher in the undispersed than in the dispersed grains (e.g. compare 0.70 and 0.75 μm grains with 10 ppm helium). Finally, it should be noticed that the linear part of the curves in Fig. 3 (i) extends to higher doses and (ii) its slope becomes smaller as void concentration decreases either on decreasing the grain size or the helium content (see Table 3, and ref. [8], Figs. 5 and 6).

4. DISCUSSION

4.1. Void Nucleation

It is generally believed that gas atoms are necessary to stabilize void nuclei. However, in order that these nuclei can survive and grow into observable voids it is equally essential that there exists a supersaturation of vacancies so as to ensure a net flux of vacancies to the void nuclei. It is expected therefore that the scale of void nucleation would be controlled by both the amount of gas atoms and the level of vacancy supersaturation. Thus it follows that for a given concentration of gas atoms, the level of void nucleation should decrease on decreasing the vacancy supersaturation. Since vacancy supersaturation is found to decrease with decreasing grain size [10], the scale of void nucleation should decrease with decreasing grain size; this is precisely what we observe experimentally (see Table 3 and ref. [8]). For a given vacancy supersaturation (i.e. grain size) the void concentration should increase on increasing the concentration of gas atoms. We do indeed observe

this kind of behavior in the grains with $d_g > 2.50$ μm (see Table 3). If it is assumed that the vacancy supersaturation in large grains is not altered by increasing the gas content, then the increase in void concentration with increasing gas content would suggest that the critical nucleus size decreases with increasing gas content; this is similar to the effect of vacancy supersaturation as discussed in ref. [8].

In the presence of very low supersaturations even very high concentrations of gas atoms do not seem to be sufficient to nucleate as many voids as in the case of large grains with high vacancy supersaturations. This implies that as a result of a decrease in the vacancy supersaturation the corresponding increase in the critical nucleus size is far greater than can be counterbalanced by increasing the gas concentration. This emphasizes the effect of vacancy supersaturation on the void nucleation.

On the basis of the above discussion it can be concluded therefore that the grain size dependencies of void nucleation and void concentration are mainly due to the reduction in vacancy supersaturation which occurs on decreasing the grain size; the effect of vacancy supersaturation is maintained even in the presence of high concentrations of helium atoms.

4.2. Dose Dependence of Void Growth and Swelling

The effect of grain size dependent vacancy supersaturation on the void volume swelling can be further examined in terms of its effect on the dose dependencies of void growth and swelling. From steady state diffusion theory Greenwood, Foreman and Rimmer [13] have shown that, neglecting the effect of thermal vacancies, the growth rate of a small spherical void of radius r is proportional to the vacancy supersaturation; their expression can be re-written in a simplified form as

$$\frac{dr}{dt} \propto \frac{1}{r} \cdot S \qquad (1)$$

where S is the vacancy supersaturation. This can be used as a basis for qualitative comparisons of the growth rates of voids in our grains of different sizes, particularly at low doses where the defect denuded zones around the voids are expected to be reasonably well separated (see ref. [14]). From eqn. (1) the dose dependence of void radius can be obtained as

$$r \propto S^{1/2} \cdot (\phi t)^{1/2} \qquad (2)$$

where ϕ is the displacement damage rate and t is the irradiation time. The dose dependence of void volume swelling follows immediately from eqn. (2), and can be written as

$$(\Delta v/v) \propto S^{3/2} \cdot (\phi t)^{3/2} \qquad (3)$$

In eqns. (1-3) the vacancy supersaturation is assumed to remain constant during the irradiation. This seems to be quite reasonable particularly in view of our observations in the High Voltage Electron Microscope that after an initial period there is generally no appreciable change in dislocation density during further irradiation.

Void Growth

The square-root dependence of void growth on displacement dose is borne out reasonably well by our results shown in Figs. 1 and 2. In grains with high void concentrations the results

begin to deviate from linearity at or beyond the dose at which C_v^{max} is reached. In the large grain containing 1000 ppm helium (Fig. 1, curve 5) the deviation seems to occur just before the void maximum is reached. This may reflect the fact that at such a high void concentration the void spacing becomes so small that the spheres of influence of the individual voids begin to overlap. In cases where void concentration is low, no deviation from linearity is observed (Figs. 1 and 2) in the dose range studied.

The fact that the void growth rate decreases with decreasing grain size (at a similar void concentration level) would, according to eqn. 2, suggest that this reduction in growth rate must be caused by a corresponding decrease in the vacancy supersaturation. A decrease in the vacancy supersaturation on decreasing the size of the grain is expected from the defect depletion model [10]. It should be noticed however that the effect of grain size on void growth is not as strong as its effect on void concentration (see Table 3). This would suggest that the grain size effect is predominantly nucleation controlled.

The dependence of void growth rate on the void concentration, as observed in large as well as small grains can be easily understood in terms of the vacancy flux available to individual voids.

The effect of helium content on the void size at a given dose and on the void growth rate is worth noting. Our results quoted in Fig. 2 clearly indicate that the presence of 1000 ppm of helium in a 0.70 μm grain (curve 1, Fig. 2) leads to a greater void size (at a given dose) and a higher growth rate than in a 0.75 μm grain (curve 4, Fig. 2) containing only 10 ppm of helium;

the void concentration in the 0.70 μm grain is also higher than in the 0.75 μm grain. No detailed and exact mechanism for this effect is available. However, the following interpretation can be advanced. An estimate of the pressure, P, in individual voids due to 1000 ppm of implanted helium in the 0.70 μm grain shows that P approaches $2\gamma/r$ where γ is the surface energy and r is the void radius. It is quite likely therefore that the thermal evaporation of vacancies from the void surface is reduced. The flux of vacancies (corresponding to the given supersaturation and void concentration) that the void receives, on the other hand, is expected to remain at least constant. Hence an increase in the growth rate is reasonable.

Void Volume Swelling

The dose dependence, according to eqn. 3, of the void volume swelling measured in the central regions of grains is shown in Fig. 3. At higher doses the experimental results begin to deviate from linearity but only in those cases where void concentration is very high. The dose at which the deviation begins generally coincides with the dose at which C_v^{max} is reached; this is very similar to the dose dependence of the void size (Fig. 1). In small grains, however, where the void concentration is fairly small, the linearity is maintained throughout the dose range investigated.

As apparent from Fig. 3, the slope of the dose dependence curves decreases with decreasing grain size. This, according to eqn. 3, implies that the decrease in swelling is caused by a corresponding decrease in the vacancy supersaturation which is consistent with our assertion that the grain size effect occurs

because of a decrease in the vacancy supersaturation.

The effect of 1000 ppm of helium on the swelling behavior of both the large (d_g > 2.5 μm) and the small (0.70 μm) grains is very marked. This higher swelling rate might be due to a higher void growth rate, as explained in the previous section.

It is also interesting to note that the swelling behavior of the 0.70 μm dispersed grain (with 1000 ppm helium) and of the 0.70 μm undispersed grain (with only 10 ppm helium) are almost identical. This is in agreement with the point made earlier (ref. [8]) that the grain size effect is stronger in the dispersed than in the undispersed material.

5. CONCLUSIONS

The results presented and discussed in this paper show that in an austenitic stainless steel both the grain refinement and dispersions of second phase particles are effective in enhancing swelling resistance. It has been shown that the swelling resistance is maintained even in the presence of high concentrations of implanted helium atoms. This is found to be consistent with our explanation of grain size effect in terms of vacancy supersaturation. It is suggested that the nucleation of voids is critically dependent on vacancy supersaturation and that the grain size effect is predominantly nucleation controlled.

6. REFERENCES

[1] Norris, D.I.R., Radiation Effects, Vol. 14, 1972, p. 1; Vol. 15, p. 1.

[2] Pugh, S.F., Loretto, M.H., and Norris, D.I.R. (Editors), "Voids Formed by Irradiation of Reactor Materials", Proceedings of the BNES European Conference, Reading, 1971.

[3] Corbett, J.W. and Ianniello, L.C. (Editors), "Radiation Induced Voids in Metals", Proceedings of International Conference, Albany, 1971.

[4] Arsenault, R.J. (Editor), "Defects and Defect Clusters in B.C.C. Metals and Their Alloys", Proceedings of International Conference, Gaithersburg, 1973.

[5] Harkness, S.D. and Li, Che-Yu, Metallurgical Transactions, Vol. 2, 1971, p. 1457.

[6] Singh, B.N., Journal of Nuclear Materials, Vol. 46, 1973, p. 99.

[7] Singh, B.N., Nature Physical Science, Vol. 244, 1973, p. 142.

[8] Singh, B.N., Philosophical Magazine, Vol. 29, 1974, p. 25.

[9] Singh, B.N., Philosophical Magazine, Vol. 28, 1973, p. 1409.

[10] Singh, B.N. and Foreman, A.J.E., Philosophical Magazine, in press.

[11] Makin, M.J., "The use of the Harwell High Voltage Microscope for Quantitative Void Studies", AERE-R-6957, A.E.R.E. Harwell, Berkshire, England, November 1971.

[12] Makin, M.J., Jernkontorets Annaler, Vol. 155, 1971, p. 509.

[13] Greenwood, G.W., Foreman, A.J.E. and Rimmer, D.E., Journal of Nuclear Materials, Vol. 4, 1959, p. 305.

[14] Foreman, A.J.E., "A Note on the Dose Dependence of Void Growth in Irradiated Materials", AERE-R-6358, A.E.R.E. Harwell, Berkshire, England, June 1970.

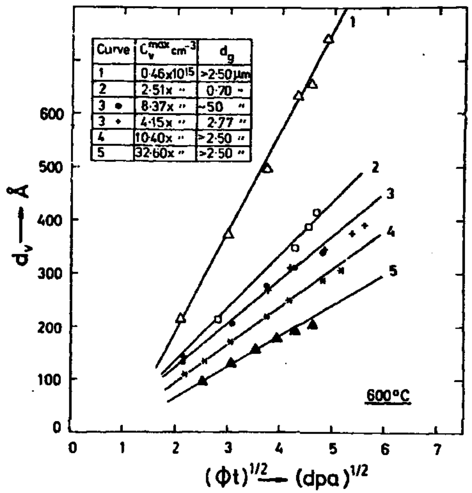

Fig. 1. Influence of grain size and helium concentration on the dose dependence of average void size in specimens without dispersions of oxide particles and irradiated at 600°C; helium concentration in ppm:

Δ	RMS-1c, 0 ppm	+	RMS-1b, 10 ppm
□	RMS-1a, 10 "	*	RMS-1c, 100 "
•	Type 316, 0 "	▲	RMS-1c, 1000 "

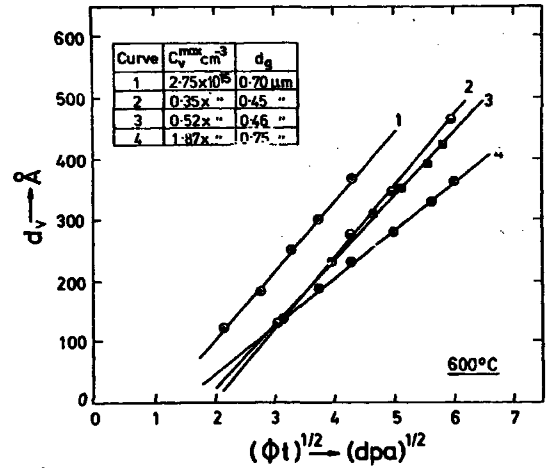

Fig. 2. Same as in Fig. 1 but with dispersions of oxide particles; helium concentration in ppm:

⊕	RMS-2a, 1000 ppm	⊞	RMS-2a, 10 ppm
⊖	RMS-2a, 0 "	⊛	RMS-2a, 10 "

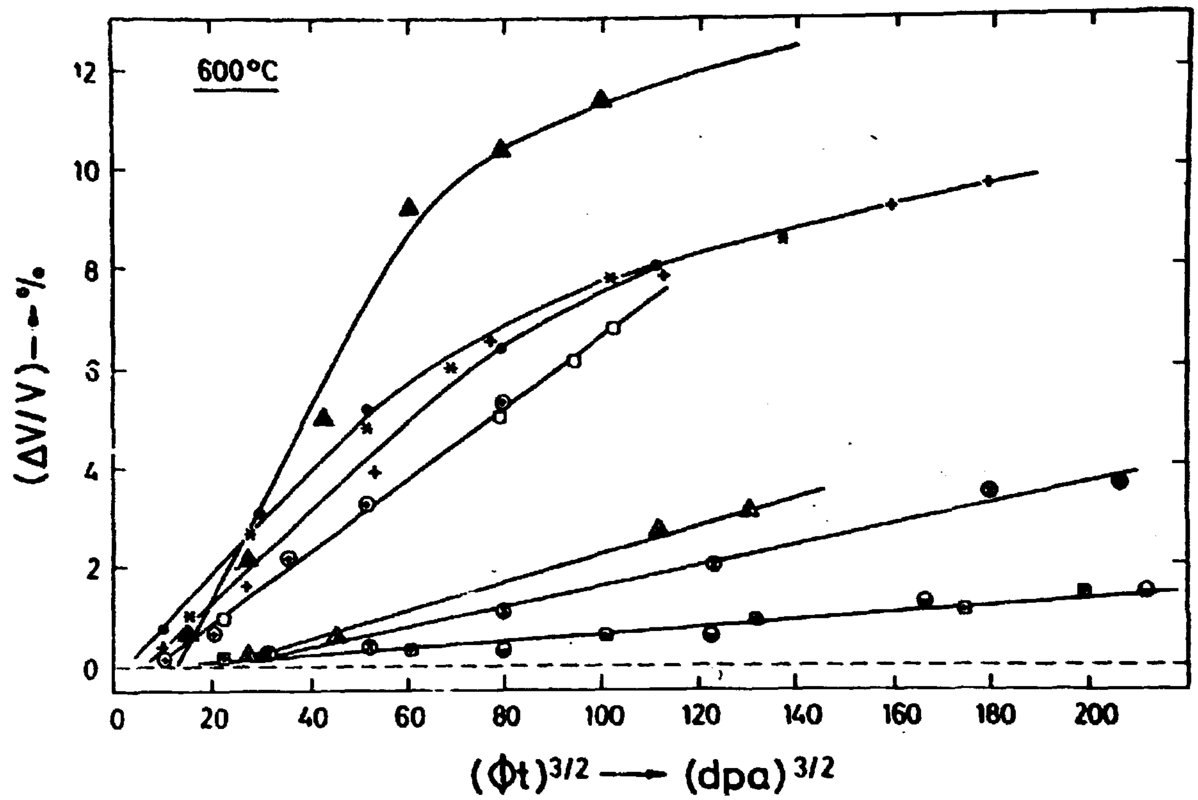

Fig. 3. Influence of grain size and helium concentration on the dose dependence of void volume swelling in dispersed and undispersed specimens irradiated at 600^{o}C; d_g in μm and helium concentration in ppm:

▲ RMS-1c, > 2.5 μm, 1000 ppm

● Type 316, ∿50 " , 0 "

* RMS-1c, > 2.5 " , 100 "

+ RMS-1b, 2.77 " , 10 "

⊕ RMS-2a, 0.70 " , 1000 "

△ RMS-2a, 0.56 " , 50 "

⊗ RMS-2a, 0.75 " , 10 "

⊞ RMS-2a, 0.46 " , 10 "

◒ RMS-2a, 0.45 " , 0 "

□ RMS-1a, 0.70 " , 10 "